Photoemission study of the diluted ferromagnetic semiconductor Zn$_{1-x}$Cr$_x$Te

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Ferromagnetic (FM) semiconductors have become one of the key materials for ‘spintronics’ devices, which aim at the integrated use of spins and carriers in semiconductors. From the application viewpoint, diluted magnetic semiconductor (DMS) showing high Curie temperature ($T_C$) and semiconducting transport properties is desired. Recently, Saito et al. succeeded in the fabrication of DMS Zn$_{1-x}$Cr$_x$Te with $T_C$ as high as 300 ± 10 K [1]. It is the highest $T_C$ ever reported for FM DMSs showing large magnetic circular dichroism [2]. Furthermore, Zn$_{1-x}$Cr$_x$Te is highly resistive [3], suggesting the controllability of transport properties in the FM phase. The mechanism of ferromagnetism in Zn$_{1-x}$Cr$_x$Te is, however, completely open, particularly because its carrier concentration of ~1×10^{15} cm$^{-3}$ [3] is 3-5 orders smaller than those of the typical III-V-based DMSs Ga$_{1-x}$Mn$_x$As and In$_{1-x}$Mn$_x$As where carrier-induced ferromagnetism [4] is proposed as the origin of ferromagnetism.

In order to study the electronic structure of Zn$_{1-x}$Cr$_x$Te, we have performed Cr 3$p$-3$d$ resonant photoemission spectroscopy (RPES) measurements on Zn$_{0.957}$Cr$_{0.043}$Te ($T_C$ ~ 70 K) and ZnTe thin films. Samples were prepared by molecular-beam epitaxy method as reported elsewhere [1,3]. Photoemission measurements were performed at BL-18A of Photon Factory. The energy resolution of the spectrometer including temperature broadening was ~300 meV. Repeated Ar-ion sputtering at 1.0 kV and subsequent annealing up to 200°C were performed to obtain clean surfaces. Surface cleanliness was checked by the weakness of the O 1$s$ and C 1$s$ core-level signals. Measurements were performed at room temperature for Zn$_{0.957}$Cr$_{0.043}$Te, and at an elevated temperature of ~450 K for ZnTe in order to minimize charging effects.

Figure 1(a) and (b) show the valence-band spectra of Zn$_{0.957}$Cr$_{0.043}$Te and ZnTe, respectively, in the Cr 3$p$-3$d$ absorption region (on resonance; $h\nu$ ~ 48 eV). The Te 4$d$ core-level structures due to second order light which were superposed on the valence-band spectra taken below $h\nu$ = 44 eV had been subtracted. One can see a large spectral intensity in the upper region of the valence band in Zn$_{0.957}$Cr$_{0.043}$Te [Fig. 1(a)] compared to that of ZnTe [Fig. 1(b)]. The cross-section of Cr 3$d$ is one order of magnitude larger than that of Te 5$p$ in the Cr 3$p$-3$d$ absorption region (26 times larger at $h\nu$ = 50 eV [5]), and therefore, we conclude that the main structure of Cr 3$d$ electrons resides in the upper region of the host valence band. Indeed one can see a clear Cr 3$p$-3$d$ resonance enhancement around the binding energy ($E_B$) of 1.7 eV. Careful examination of the constant-initial-state spectra (not shown) indicated another Cr 3$p$-3$d$ resonance enhancement at higher $E_B$ region of ~ 8 eV, This structure is assigned to the bonding state of Cr 3$d$ and neighboring Te 4$p$ orbitals. In the LDA calculations, Zn$_{1-x}$Cr$_x$Te is predicted to be a half-metallic ferromagnet, but it is clear that the spectral intensity around the Fermi level is strongly suppressed, consistent with its semiconducting behavior.


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